

minimize potential electron-beam effects, the authors ran the full deformation cycle with the electron beam off, and turned it on at the end of the process to image the final shape, which they showed to be identical to that before the deformation process started. They also show that, at low current, heating effects are small. Although these experiments do not definitely rule out all potential electron-beam effects on the surface-diffusion process, the authors have taken great care to reduce the beam current to values that are low enough for the influence of the beam on surface diffusion to be negligible⁷.

Li and colleagues' findings have consequences for basic science and for the practical use of nanocrystals — for example, as used in nanoelectronic devices. On the

one hand, their experimental approach should make possible the systematic study of the relaxation time of nanocrystals of different sizes as they recover their equilibrium shape at room temperature. Such a study would serve as a test of the power-law dependence of relaxation time on nanocrystal size¹. On the other hand, nanocrystals can temporarily suffer stresses that irreversibly change the nanocrystal's shape, potentially causing device failure. Also, nanocrystal shape can evolve rapidly under varying chemical conditions, such as changes in the composition of its immediate environment. This could cause device damage but could also be an advantage in gas-sensing devices, as shape changes induced by gas adsorption may cause changes in electrical conductivity.

All in all, Li and colleagues' results highlight that crystals at the nanoscale can deform like liquids. □

Claude R. Henry is at the Centre Interdisciplinaire de Nanoscience de Marseille, Aix-Marseille Université / CNRS, UMR 7325, F-13288 Marseille cedex 09, France.
e-mail: henry@cinam.univ-mrs.fr

References

1. Sun, J. *et al.* *Nature Mater.* **13**, 1007–1012 (2014).
2. Herring, C. in *The Physics of Powder Metallurgy* (ed. Kingston, W. E.) 143–178 (McGraw Hill, 1951).
3. Giorgio, S. *et al.* *Ultramicroscopy* **106**, 503–507 (2006).
4. Cabié, M. *et al.* *J. Phys. Chem. C* **114**, 2160–2163 (2010).
5. Yoshida, H. *Appl. Phys. Express* **4**, 065001 (2011).
6. Giorgio, S., Cabié, M. & Henry, C. R. *Gold Bulletin* **41**, 167–173 (2008).
7. Wang, Z. W. & Palmer, R. E. *Nano Lett.* **12**, 91–95 (2012).
8. Surrey, A., Pohl, D., Schultz, L. & Rellinghaus, B. *Nano Lett.* **12**, 6071–6077 (2012).

ELECTRON MICROSCOPY

Shape of a crystal from one image

Aberration-corrected electron microscopes are now being exploited to achieve quantitative atomic-resolution information about surface morphology from a single image.

Leslie J. Allen

In 1994, Hÿtch and Stobbs¹ pointed out the existence of a large contrast mismatch between high-resolution electron microscopy images that were obtained using conventional transmission electron microscopy (CTEM) and simulations, a mismatch known as the Stobbs factor. This was also the case for images obtained with scanning transmission electron microscopy (STEM), where a fine probe is raster-scanned across the specimen to form an image. Despite the substantial improvements in resolution offered by the advent of aberration-corrected electron microscopy in the years following², it was not until 2008 in the case of STEM³ and 2009 for CTEM⁴ that quantitative imaging was demonstrated, overcoming the problems related to the Stobbs factor. This quantification capability in STEM imaging has led to counting of atoms in a gold foil⁵ and to locating impurity atoms in three dimensions^{6,7}. Now, a further major advance in quantitative imaging has been made using CTEM. Writing in *Nature Materials*⁸, Jia and co-workers demonstrate the experimental reconstruction of the three-dimensional (3D) shape of a thin magnesium oxide crystal with single-atom precision from only one image. The reconstruction

procedure is able to reveal the surface morphology of the crystal with atomic resolution with the benefit of detecting adsorbed impurity atoms.

The approach used by Jia and co-workers differs fundamentally from the standard 3D reconstruction approach used in tomography, which relies on the combination of the information gathered by different views (or 2D projections) of the object being imaged. The difference is that Jia and colleagues make efficient use of the fact that electron diffraction is intrinsically a 3D process. Figure 1 illustrates⁸ how this 3D information can be used: a magnesium dioxide crystal is illuminated from the top by the CTEM electron beam. The structural information about the distribution of atoms in each column of the crystal is encoded in how the illumination above that atomic column interacts with it, leading to subtle variation in contrast as one moves from column to column (Fig. 1, labels i, j). The instrumentation produced TEM images with an absolute greyscale and a good signal-to-noise ratio. The spatial variations in the images were then compared with simulations. A structure refinement iterative procedure was used to find the best fit between a structure model and the experimental image. A

statistical confidence check provided precise quantitative statements about the uniqueness of the final 3D structure model. In this way it was possible to gather information on column positions, number of atoms in a column and the species of the top-most atom, as well as on how a column is located along the optical axis, thus determining the surface morphology of the crystal (Fig. 1). In fact, it was even possible to identify sites that appeared 'half-occupied' (brown and cyan in Fig. 1), most likely due to impurity carbon atoms. Additionally, the results show that single-atom sensitivity is not only obtainable for high nuclear charge elements but also for light chemical elements, such as oxygen. This is of considerable practical relevance because oxygen plays a key role as an anionic constituent in many technologically important materials.

Several factors contributed to the success of the approach used by Jia and colleagues⁸. Carefully chosen imaging conditions were used to provide strong localized image contrast. A good signal-to-noise ratio in the recorded image data was essential to clearly distinguish between candidate structures that, as can be seen in simulations, have small differences in intensity. These imaging conditions require

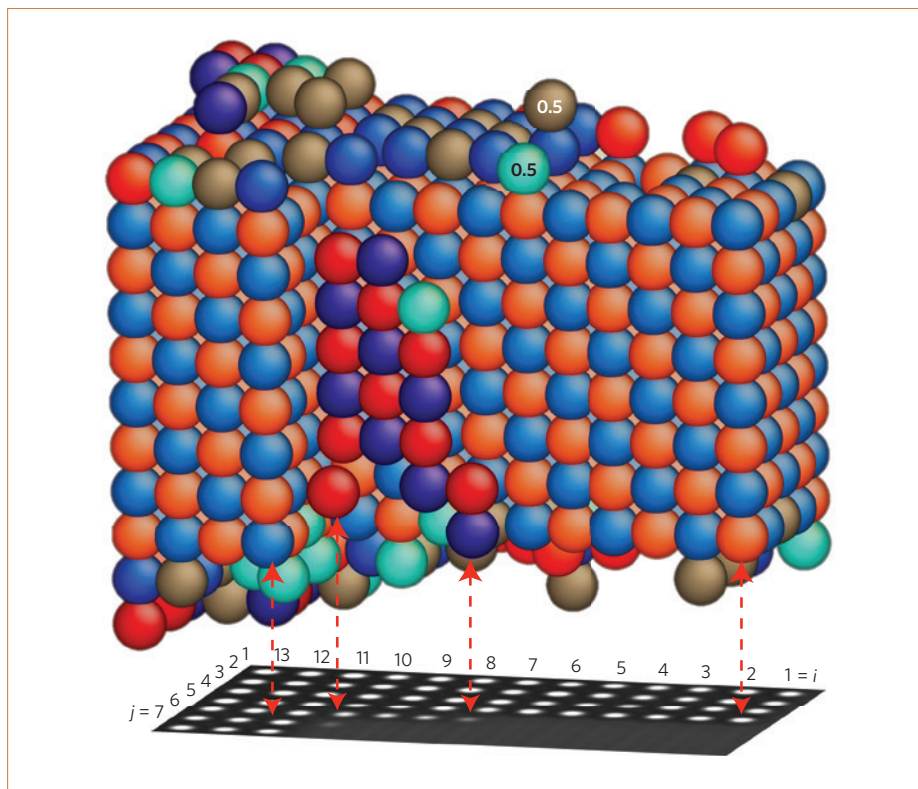


Figure 1 | The arrangement of atoms in three dimensions in a magnesium oxide nanocrystal⁸. Red and blue spheres denote fully occupied Mg and O sites, respectively. The increased colour saturation highlights surface atoms, that is, atoms are indicated in a darker shade of red and blue on the top, bottom and side surfaces. Brown and cyan spheres denote half-occupied Mg and O surface sites, respectively.

a microscope with excellent electronic and mechanical stability, good spatial and temporal coherence properties of the electron beam and a sensitive camera. A crystalline structure free of impurities or

vacancies was used so that the analysis of the data could correctly assume a discrete lattice of atoms, disallowing continuous intermediate z positions. A suitably large difference in the atomic charge numbers of

the constituents of magnesium oxide ($Z = 8$ for O, and $Z = 12$ for Mg) was also a factor in the success of the analysis. The result highlighted here was made possible by a quantitative approach to image analysis and by achieving agreement between experiment and simulation. In this way, the work by Jia and co-authors contributes significantly to the quest for quantitative atomic-resolution electron microscopy^{9,10} and will have an impact in both the materials and biological sciences. It is likely that the structural sensitivity of the method can be enhanced significantly by modified experimental conditions such as by using, for example, low-voltage microscopy, cooling stages or ultrahigh-vacuum conditions. The ability to determine 3D information from a single image will be particularly pertinent for beam-sensitive specimens, such as biological crystals, where it is important to keep the electron dose low. □

Leslie J. Allen is in the School of Physics, University of Melbourne, Victoria 3010, Australia.
e-mail: lja@unimelb.edu.au

References

1. Hýtch, M. J. & Stobbs, W. M. *Ultramicroscopy* **53**, 191–203 (1994).
2. Urban, K. W. *Nature Mater.* **8**, 260–262 (2009).
3. LeBeau, J. M., Findlay, S. D., Allen, L. J. & Stemmer, S. *Phys. Rev. Lett.* **100**, 206101 (2008).
4. Thust, A. *Phys. Rev. Lett.* **102**, 220801 (2009).
5. LeBeau, J. M., Findlay, S. D., Allen, L. J. & Stemmer, S. *Nano Lett.* **10**, 4405–4408 (2010).
6. Hwang, J., Zhang, J. Y., D'Alfonso, A. J., Allen, L. J. & Stemmer, S. *Phys. Rev. Lett.* **111**, 266101 (2013).
7. Ishikawa, R., Lupini, A. R., Findlay, S. D., Taniguchi, T. & Pennycook, S. J. *Nano Lett.* **14**, 1903–1908 (2014).
8. Jia, C. L. *et al. Nature Mater.* **13**, 1044–1049 (2014).
9. Xin, H. L., Dwyer, C. & Muller, D. A. *Ultramicroscopy* **139**, 38–46 (2014).
10. Kothleitner, G. *et al. Phys. Rev. Lett.* **112**, 085501 (2014).

TRIPLET EXCITONS

Bringing dark states to light

Semiconducting quantum dots have been used to harvest triplet excitons produced through singlet fission in organic semiconductors. These hybrid organic–inorganic materials may boost the efficiency of solar cells.

Christopher J. Bardeen

A goal of current research in solar energy is to develop strategies for making so-called third-generation solar cells that exceed the Shockley–Queisser limit on photovoltaic efficiency¹. According to this limit, nearly two-thirds of the light energy incident on a traditional photovoltaic material is not converted to electrical energy. This is partly due to the rapid relaxation of

excitons generated by high-energy photons to the lowest available energy states (corresponding to the bandgap energy of the material), with a consequent loss of the excess photon energy as waste heat. If a photon at twice the bandgap energy could be converted into a pair of low-energy excitons, then its excess energy could generate extra photocurrent and boost the overall solar conversion efficiency by as

much as 30%². This down-conversion can be done using glasses containing rare earth ions³, where it is also known as ‘quantum cutting’, but the materials are expensive and tend to operate over narrow wavelength ranges. In organic semiconductors — which have the potential to be manufactured and processed into devices at low cost — this conversion process is known as singlet fission (SF), in which a photon creates a